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FINAL REPORT

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Photoemission, Vibrational and Stimulated Desorption Studies of Metal-Semiconductor Interfaces and of Chemisorbed Atoms and Molecules

Principal Investigator: Giorgio Margaritondo
Department of Physics, University of Wisconsin,
Madison, WI 53706
(Phone: (608) 263-3919 or (608) 873-7722)

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Results Obtained by the Program

This program has produced a large number of results, reported in many articles and invited presentations. The quality of the production is as good as its quantity, as demonstrated by the highlights discussed in this section.

Most of the past results of the program have been obtained on metal/semiconductor interfaces. However, the recent months have brought exciting new developments in the field of high-temperature superconductors. We submitted two articles describing our first results in the latter field, sixty days after we learned about superconductivity above 90K.

Metal-Semiconductor Interfaces

Three series of results recently produced by this program are particularly relevant to the development of a general, "unified" theory of metal-semiconductor interfaces:

1. The study of Al overlayers on Si(111) by photoemission and high-resolution energy loss spectroscopy, which established a link between the appearance of local metallic character and the formation of the Schottky barrier.[1]
2. The study of metal chemisorption on GaAs substrates as a function of the substrate temperature in the range from liquid nitrogen (LNT) to room temperature (RT). This study revealed the interplay of different factors which must be considered in a "unified" description of the Schottky barrier formation process. In some cases, it revealed again a correlation between the final establishment of the Schottky barrier and the appearance of local metallic character.[2]
3. The first study of a semiconductor overlayer on a single-crystal metal substrate, Si on Al(111), and in particular the discovery of the 3×3 reconstruction of the Si overlayer at submonolayer coverages. These experiments, in our opinion, were long overdue. In fact, literally hundreds of experiments have explored metals on semiconductors, while to our knowledge there were no previous studies of semiconductors on single-crystal metals.[3]

In order to understand the significance of the above results, and in particular of the last

[1] M. K. Kelly, E. Colavita, G. Margaritondo, L. Papagno, J. Anderson, D. J. Frankel and G. J. Lapeyre, Phys. Rev. **B32**, 2693 (1985); M. K. Kelly, G. Margaritondo, J. Anderson, D. J. Frankel and G. J. Lapeyre, J. Vac. Sci. Technol. **A4**, 1396 (1986); M. K. Kelly, G. Margaritondo, L. Papagno and G. J. Lapeyre, Phys. Rev. **B34**, 6011 (1986).

[2] M. K. Kelly, A. Kahn, N. Tache, E. Colavita and G. Margaritondo, J. Vac. Sci. Technol. **A4**, 882 (1986); M. K. Kelly, A. Kahn, Nacira Tache, E. Colavita and G. Margaritondo, Solid State Commun. **58**, 429 (1986).

[3] Y. Chang, E. Colavita and G. Margaritondo, to be published. Also see: A. Franciosi, D. W. Niles, G. Margaritondo, C. Quaresima, M. Capozi and P. Perfetti, Phys. Rev. **B32**, 6917 (1985), and D. W. Niles, N. Tache, D. G. Kilday and G. Margaritondo, Phys. Rev. **B34**, 967 (1986).

one, it is necessary to consider our present status of knowledge of metal-semiconductor interfaces, and of semiconductor interfaces in general. Before the advent of surface-sensitive experiments, the models used to explain Schottky barrier relied on a simplistic description of the local electronic structure of the interface. The most advanced descriptions were Bardeen's model, based on the pinning of the Fermi level by surface/interface states, and Heine's model, based on the effect of tunneling of the metal wave functions into the semiconductor gap.

Surface-sensitive experiments demolished some of the most simplistic notions, such as the attribution of the Fermi-level pinning to the *clean* surface states of the semiconductor. These experiments also revealed a number of phenomena and factors that could, at least in principle, influence the Schottky barrier height. For a certain period of time, the situation seemed hopelessly complex — each interface appeared as a "special case". Eventually, however, from the analysis of a wide array of experimental results certain factors emerged as the leading candidates in the establishment of the Schottky barrier. Landmarks in this process were the formulation of the *unified defect model*, the work on the concept of *metal-induced gap states* (MIGS), and the introduction of the *effective work function model.*

In the first case, localized defects created as a by-product of the interface formation process pin the Fermi level at fixed energies in the gap of the semiconductor, giving the Schottky barrier height. In the second case, the Schottky barrier height is determined by aligning with the metal Fermi level the "midgap-energy point" or "charge-neutrality point", i.e., the energy at which the MIGS character changes from metal-like to conduction-like. In the effective work function model, the Schottky barrier is established by a Schottky-like mechanism, in which the work function of the metal is replaced by that of the local chemical species formed at the interface.

The merits and demerits of the above three points of view have been the subject of intense controversy for the past several years. Our recent results show that none of these approaches can explain *all* Schottky barrier properties. Consider, for example, the case of metal-GaAs interfaces. For RT substrates, Spicer and co-workers provided good evidence in favor of the "unified" defect model. However, we have shown that the behavior is totally different for different substrate temperatures, and it depends on the substrate doping. The defect model can be amended to introduce more and more defects and/or defect levels, and explain the increased complexity. This, however, is not the spirit of a *unified* defect model. Furthermore, our temperature-dependent data revealed, in some cases, a correlation between the appearance of metallic character at the interface and the final establishment of the Schottky barrier, as predicted by the MIGS model rather than by the defect model.

Similar evidence in favor of the MIGS model is provided by our detailed studies of the prototypical interface obtained by depositing Al on Si. The clean Si(111)7×7 substrate has metallic character, and there is evidence that such a character is due to a narrow band of states at Fermi level, located inside a small gap.[4] We find that the metallic character (detected by studying the elastic-peak lineshape in high-resolution electron energy loss) disappears when the first Al adatoms are deposited, probably due to the removal of the narrow band. Furthermore, we found that the width of the gap increases. Thus, metal adatoms on this semiconductor surface transforms a local metal into a local insulator!

As the overlayer thickness increases, the metal character reappears at a coverage of the order of 1.5 monolayer, the same coverage at which the motion of the Fermi level in the gap ends, and the Schottky barrier is formed. This is entirely consistent with the predictions of MIGS models. One should not, however, jump to the conclusion that this approach works for *all* Si-Al interfaces. In fact, when we deposit Si on Al, we find evidence of a local electronic structure similar to that of Al on Si. However, the Fermi-level pinning positions are different in the two cases, indicating that a MIGS mechanism determines the Schottky barrier in one case but not in the other.

The third theoretical point of view, the effective work function model, predicts a linear dependence of the Schottky barrier height on the work function of the metallic interface species. Such a dependence *is* experimentally observed. Furthermore, it appears more marked in the case of large-gap semiconductors. For example, we found that metal-GaP interfaces behave as almost perfect Schottky systems, without even invoking the "effective" work functions.[5]

These results are not necessarily inconsistent with the MIGS approach and with the defect model. In fact, the most recent versions of these theories include a Schottky-like term. The role of this term depends on the screening, which in turn depends on a suitable dielectric constant and therefore on the magnitude of the gap. It is not surprising, therefore, that large-gap semiconductors such as GaP tend to exhibit a Schottky-like behavior. The description of the effects of screening on the Schottky term are qualitatively successful, although quantitatively inadequate because of the difficulty in using dielectric constants for these heterogeneous and inhomogeneous systems.[6]

In our opinion, the message from the above data is quite clear. The three factors treated by the above approaches are all potentially important in determining the Schottky barrier height. Specifically, for low interface defect density, the MIGS factor prevails, with an

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[4] J. E. Demuth, B. N. J. Persson and A J. Schell-Sorokin, Phys. Rev. Lett. **51**, 2214 (1983).

[5] L. J. Brillson, R. E. Viturro, M. L. Slade, P. Chiaradia, D. Kilday, M. K. Kelly and G. Margaritondo, Appl. Phys. Lett. **50**, 1379 (1987); P. Chiaradia, L. J. Brillson, M. Slade, R. E. Viturro, D. Kilday, N. Tache, M. K. Kelly and G. Margaritondo, J. Vac. Sci. Technol. (in press).

[6] J. Tersoff and G. Margaritondo, unpublished.

additional Schottky term whose importance depends on the screening. For high interface defect density, the defect factor prevails, again with an additional Schottky term. Thus, the Schottky barrier for p-type materials can be written, in general:

$$\Phi_{B,p} = \alpha E_B + \beta E_d + f(E_g)\Delta\chi,$$

where E_B is the semiconductor midgap-energy point (measured from the top of the valence band), which defines the Schottky barrier in the MIGS model; E_d is the Fermi-level pinning position at a defect level; the coefficients α and β depend on the local defect density; the term $\Delta\chi$ is the Schottky term, *i.e.*, the difference between the metal work function (or the effective work function of the interface species) and the semiconductor electron affinity corrected for the gap; $f(E_g)$ is a decreasing function of the gap, E_g, which describes the effects of screening. The extension to the n-type case is trivial.

This "unified" hypothesis is supported by the results of our program. For example, the dependence of the Si-Al Schottky barrier on the deposition sequence is explained by assuming that MIGS prevail when Al is deposited on Si — while the deposition of Si on Al creates a large density of acceptor-like vacancies in the overlayer which pin the Fermi close to the top of the valence band. Evidence in favor of this approach was recently presented by Mönch,[7] who demonstrated that most Si Schottky barriers can be assigned to either one of two classes, one consistent with the defect model and the other with the MIGS model.

We emphasize that the present evidence is not sufficient to demonstrate that this "unified" approach is valid for all Schottky barriers, or even for a large portion of Schottky barriers. Extensive experimental studies are necessary. We do believe, however, that this hypothesis should be the guideline in the future development of metal-semiconductor interface research, and we developed our plans accordingly.

The interest in metal-semiconductor interfaces has been enhanced by a recent, exciting discovery — there is a clear correlation between Schottky barrier heights and heterojunction valence band discontinuities.[8] Such a correlation is predicted, for example, by the Schottky model and its heterojunction counterpart, Anderson's electron affinity rule.[9] In its simplest form, the predicted correlation is that the difference of the p-type Schottky barrier heigths for two semiconductors and the same metal equals the valence-band discontinuity for the heterojunction interface between the same two semiconductors. We used our extensive data set on heterojunctions to prove that such a correlation does indeed exists. This point is discussed in a recent article we published in Physics Today,[10]

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[7]See: W. Mönch, Physical Review Lett. **58**, 1260 (1987), and references therein.

[8]D. W. Niles, G. Margaritondo, E. Colavita, P. Perfetti, C. Quaresima and M. Capozi, J. Vac. Sci. Technol. **A4**, 962 (1986); also see: F. Capasso and G. Margaritondo: *Heterojunctions: Band Discontinuities and Device Applications* (North Holland, Amsterdam, in press).

[9]R. L. Anderson, Solid State Electron. **5**, 341 (1962).

[10]R. S. Bauer and G. Margaritondo, Physics Today **40**, 27 (1987).

It can be shown that, under certain assumptions, the above correlation is also consistent with the MIGS model and with the defect model — and, therefore, with the "unified" hypothesis based on their interplay. This means that our results in testing the "unified" hypothesis for Schottky barriers will have a strong impact on heterojunction research, and vice-versa. The implications are really interesting. For example, we demonstrated, in two independent series of experiments,[11] that thin inteface intralayers can change Schottky barrier heights as well as heterojunction band discontinuities. Thus, the results of this research could teach us how to "tailor" the most important interface parameters, and enhance the performances and the flexibility of a wide variety of semiconductor devices.

The research along the guidelines suggested by the "unified" hypothesis must be conducted primarily on simple interfaces. We will not, however, neglect the study of more complex, and perhaps more "real", interfaces. These investigations have produced many interesting results in recent years. For example, in collaboration with Martin-Marietta Laboratories we have clarified many fundamental aspects of the local stoichiometry of interfaces involving the technologically important narrow-gap semiconductor (Hg,Cd)Te.[12] The most important result of these studies is that the interface formation process often causes large deviations in the local stoichiometry from the desired compositions so painfully obtained by the crystal growers. These deviations drastically change the characteristics of the corresponding devices. The continuing research in this area is producing extensive and complete data, which will make it possible to understand the factors which determine the changes in local stoichiometry.

Another productive area of our program is the study of interfaces involving ternary III-V materials.[13] These samples are produced with advanced molecular beam epitaxy (MBE) methods by Woodall and co-workers at IBM, then passivated, transferred to one of our spectrometers at SRC, cleaned *in situ* and investigated with synchrotron radiation photoemission. The experiments, conducted with Len Brillson (another ONR contractor) and co-workers, have produced the first data on interface formation on these materials, revealing differences and analogies with respect to their binary counterparts.

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[11] C. F. Brucker, L. J. Brillson, A. D. Katnani, N. G. Stoffel and G. Margaritondo, J. Vac. Sci. Technol. **21**, 590 (1982).

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High-Temperature Superconductors

Our interest in superconductors was, of course, stimulated by the recent, exciting discoveries in this field. This research is also linked to studies we conducted for several years on layered materials, and in particular on transition-metal dichalcogenides.[14] The charge-density-wave phase transitions observed in the latter have several points in common with some of the mechanisms which have been proposed for high-temperature superconductivity in copper oxide materials.

A few years ago, we presented the first and only – to our knowledge – experimental observation of the modifications in the density of states near the Fermi level due to the opening of an instability gap.[15] The results were obtained for the charge-density-wave-like transition of $TiSe_2$, a proposed "excitonic insulator". These experiments were made easy by the magnitude of the density-of-state changes, which in turn corresponds to an unusually large instability gap. Our preliminary tests on $Ba_2YCu_3O_{7-x}$ indicate[16] a much smaller instability gap, consistent with the results of other groups. However, it may still be possible to observe the corresponding small changes in the density of states, as discussed in the next section.

We performed extensive studies of $Ba_2YCu_3O_{7-x}$ with synchrotron radiation photoemission, electron energy loss and Auger spectroscopy. The experiments were performed on materials grown and characterized by Tarascon and co-workers at Bell Core and by Hellstrom, Larbalestier and co-workers at Wisconsin. The electron spectroscopy experiments were performed in collaboration with M. Onellion, and intepreted in collaboration with Robert Joynt, both of Wisconsin. The results provide a rather complete and consistent picture of the electronic structure of this material in a range from 40 eV below to 6 eV above the Fermi level.

The importance of using synchrotron radiation for these experiments is determined, in particular, by the occurrence of strong *photoemission resonances*, observed when taking photoemission spectra at different photon energies.[17] The intensity of a photoemission feature is strongly enhanced when the photon energy is immediately above one of the optical absorption thresholds of the atomic species which produces the feature. The enhancement is due to the quantum interference between two different processes with equal initial and final states, *i.e.*, the direct excitation of a photoelectron and the threshold optical absorption followed by de-excitation and transfer of the energy to the electron which

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[14] G. Margaritondo, in *Physics and Chemistry of Materials with Low Dimensional Structure - Electronic States and Electronic Transitions in Layer Materials*, V. Grasso Ed. (Reidel, Dordrecht 1986), Vol. 20, Chapter 6.

[15] G. Margaritondo, C. M. Bertoni, J. H. Weaver, F. Lévy, N. G. Stoffel and A. D. Katnani, Phys. Rev. **B23**, 3765 (1981).

[16] M. Onellion, Y. Chang, D. W. Niles, Robert Joynt, G. Margaritondo, N. G. Stoffel and J. L. Tarascon, to be published

[17] See, for example: G. Margaritondo, *Introduction to Synchrotron Radiation* (Oxford, New York, in press)

becomes a photoelectron. The quantum interference requires reasonable coherence, and therefore resonant phenomena are typically observed for localized f-states and d-states, and for "satellite" photoemission features.

Photoemission resonances are extremely helpful for the identification of the different contributions to the photoemission spectra. This is particularly important in the case of samples with many atomic species such as high-temperature superconductors. We used this approach to identify Ba contributions and Cu satellite contributions. With this initial clarification, it was then possible to use the photon energy dependence of the photoionization cross section to identify other Cu and O contributions.

Several points revealed by our experiments are relevant to the explanation of the superconducting mechanism. First, we agree with the theoretical predictions[18] of a low density of states near the Fermi level. Second, we did not observe temperature-induced changes in this low density of states. This indicates that the instability gap is small, contrary to what was observed in systems for which instabilities are driven by excitonic and/or charge-density-wave phenomena, such as $TiSe_2$. Third, the analysis of the resonant phenomena rules out a 1+ valence state for Cu. We are continuing the analysis to identify features which could distinguish between a 2+ and a 3+ state.

In parallel to the "conventional" photoemission studies, we also performed experiments in the "partial-yield" mode, which essentially measures the local optical absorption coefficient of the system. We also took electron-energy-loss spectra. From the parallel analysis of these two kinds of spectra, we could identify the energy of the bulk plasmon, and of another collective excitation, tentatively attributed to a surface plasmon. We could also explain the observed optical transitions in terms of two peaks in the unoccupied density of states.

[18] S. Massidda and Art Freeman, private communications

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Books and Book Chapters

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Patents Filed or Granted: N/A

Invited Presentations at Conferences

1. "Applications of Synchrotron Radiation – Semiconductors", Industrial Workshop on the 6 GeV Source, Chicago 1985.

2. "Photoemission Spectroscopy in Industrial Semiconductor Devices", Summer School on the Industrial Applications of Synchrotron Radiation International Center for Theoretical Physics, Trieste 1985. item "Photoemission Studies of Interfaces", International Conference on Synchrotron Radiation, Trieste, Italy, 1986.
3. "Overview of Synchrotron Ultraviolet Research", SURA Conference on Synchrotron Radiation Research, Atlanta, Georgia, 1986.
4. "Synchrotron Radiation Spectroscopy of Semiconductor Interfaces", Surface Canada 1986, London, Ontario, 1986.
5. "III-V Interfaces: Schottky Barriers vs. Heterojunctions", Workshop on III-V Semiconductor-Metal Interfacial Chemistry and its Effects on Electrical Properties, Stanford, CA, 1986.
6. "Synchrotron Radiation in Semiconductor Research: Fundamental and Applied Aspects", Annual Scientific Meeting of the Semiconductor Section of the GNSM, National Research Council, Rome, Italy, 1986.
7. "New Results on Metal-Semiconductor Interfaces from Energy Loss and Photoemission Experiments", 11th Annual Meeting on Advances in Surface and Interface Physics, Modena, Italy, 1986.
8. "Characterization of Semiconductor Interfaces with Synchrotron Radiation", 1987 Annual Meeting of the American Vacuum Society, Illinois Chapter.
9. "Heterojunctions and Schottky Barriers: Similarities, Differences and What We Can Learn from Them", International Workshop on Superlattice Structures and Devices, Minneapolis 1987.

Honors/Awards/Prizes: N/A

Graduate Students Supported under Contract

1. David W. Niles
2. James McKinley

Postdoctorals Supported under Contract: N/A